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- Proprietor: TORAY INDUSTRIES, INC. 2, Nihonbashi Muromachi 2-chome Chuo-ku Tokyo 103 (JP)
- (7) Inventor: Deguchi, Yukichi
  A6-23, 10, Sonoyama 2-chome
  Ohtsu-shi Shiga-ken (JP)
  Inventor: Miyaji, Shinichiro
  483 Minakuchi-cho Hatta
  Kohga-gun Shiga-ken (JP)
  Inventor: Kobayashi, Hiroaki
  B3-24, 15, Sonoyama 2-chome
  Ohtsu-shi Shiga-ken (JP)
- (7) Representative: Miller, Joseph et al J. MiLLER & CO. Lincoln House 296-302 High Holborn London WC1V 7JH (GB)

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## Description

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The present invention relates to a biaxially orientated poly-p-phenylene sulfide film. More particularly, it relates to a chemically stabilized biaxially orientated poly-p-phenylene sulfide film.

Poly-p-phenylene sulfide is a thermoplastic crystalline engineering plastic which is superior in thermal resistance, electrical insulating properties, chemical resistance, and flame retardancy. It is useful as a raw material for injection molding as well as films and fibers. Molded materials, films, and fibers produced therefrom are suitable as heatproof electrical insulating materials because of their long-term thermal resistance (continuous use at 160° to 200°C).

However, the use of poly-p-phenylene sulfide as an electrical insulating material has been limited due to the following disadvantages.

First, the conventional poly-p-phenylene sulfide insulating material is subject to so-called "copper contact failure" which is a phenomenon that the insulating material considerably decreases in thermal resistance when it is exposed to a high temperature continuously while in direct contact with copper, as in the case where an insulating film is wound directly around a copper wire.

Secondly, the conventional poly-p-phenylene sulfide insulating material decreases considerably in thermal resistance when it is exposed to a high temperature continuously in the presence of oxygen in an enclosed environment, as in the case where the insulating material is used in an enclosed motor. This phenomenon is less severe when it is used in an open motor.

Thirdly, the conventional poly-p-phenylene sulfide resin composition shortens the life of a mold due to gradual corrosion of the part of the mold in contact with the polymer during injection molding even in applications other than electrical insulation.

The above disadvantages are also found in the biaxially oriented poly-p-phenylene sulfide film for electrical insulation as disclosed in U.S. Patent No. 4,286,018. This patent discloses that the biaxially oriented poly-p-phenylene sulfide film can contain an additive and/or filler, but this disclosure merely suggests the possibility for incorporating an additive and/or filler and the incorporation of an additive and/or filler is not intended to eliminate the above-mentioned disadvantages. This patent mentions nothing about the improvement in resistance to copper contact and thermal resistance in closed systems which is accomplished by the use of specific additives or fillers described hereinafter.

There is disclosed in U.S. Patent No. 4,140,671 the addition of a metal oxide such as ZnO and MgO to a polyethylene terephthalate resin composition containing polyphenylene sulfide. According to the disclosure, the metal oxide additive is incorporated merely as a nucleating agent for the polyethylene terephthalate mainly constituting the composition and the presence of the polyethylene terephthalate as a main ingredient is a requisite of this invention. What is disclosed in this patent is not associated with the improvement in the disadvantages of poly-p-phenylene sulfide resin compositions described heretofore. This is apparent from the well known fact that a thermoplastic polyethylene terephthalate resin composition as disclosed in this patent is so poor in thermal resistance it is unable to be used for a long time under a high temperature condition where copper contact failure becomes a problem.

It is an object of the present invention to provide biaxially oriented poly-p-phenylene sulfide film which is improved in resistance to copper contact and thermal resistance in closed systems and is applicable in broad application areas as an electrical insulating material.

It is further another object of the present invention to provide a poly-p-phenylene sulfide resin composition suitable for manufacturing molded materials, fibers, nonwoven fabrics, and paper of poly-p-phenylene sulfide which are improved in resistance to copper contact and thermal resistance in closed systems.

The present invention in gist relates to a biaxially orientated poly-p-phenylene sulfide film prepared from a resin composition comprising poly-p-phenylene sulfide as a principal component of 0.05 to 40 wt% (based on the total weight of the composition) of at least one component selected from fine zinc oxide particles and fine lead oxide particles, wherein particles not larger than one µm in diameter account for at least 10% of the total number of particles.

The poly-p-phenylene sulfide (abbreviated as PPS hereinafter) as the major component of the composition of this invention should be composed of at least 90 mol%, preferably at least 95 mol%, of the recurring units represented by the structural formula

If the content of the p-phenylene sulfide units is less than 90 mol%, preferably less than 95 mol%, the resulting polymer is not sufficient in crystallinity and low in thermal deformation temperature and it is difficult to prepare superior plastic materials from such a polymer (the term "plastic materials" comprehends any product in the form of lump, sheet, and fiber made by utilizing the thermoplasticity of PPS).

The remainder, which is less than 10 mol%, preferably less than 5 mol%, of the recurring units of the polymer may contain

meta units, ( 
$$S-$$
), ether units (  $O-$ 0),

sulfone units (  $O-$ 50, biphenyl units (  $O-$ 5-),

naphtyl units (  $O-$ 5-), and substituted phenyl sulfide units (  $O-$ 5-)

(where R is an alkyl group having 1 to 10 carbon atoms, nitro group, phenyl group, or alkoxyl group having 1 to 10 carbon atoms), or

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in an amount not greatly affecting the crystallinity, stretchability, sensitivity to thermal degradation, and processability of the resulting polymer. It is specially preferable that these comonomer units is less than 5 mol% of the total recurring units.

The polymer should have an apparent melt viscosity of 5 to 10,000 Pa. sec, preferably 7 to 5,000 Pa. sec, as measured at a temperature of 300°C and under an apparent shear rate of 200 sec<sup>-1</sup> from the standpoint of processability.

The apparent melt viscosity is not directly related with the melt flow index (MFI) which is generally used as an index for melt viscosity, but the PPS composition of this invention has an MFI in the range of 3 to 6000.

The degree of polymerization of PPS is difficult to determine accurately because PPS does not dissolve at all in an usual organic solvent and it varies greatly depending on the composition of comonomers and the degree of crosslinking. It is approximately 10 to 1000.

The PPS resin composition of this invention should contain at least one component selected from fine zinc oxide particles and fine lead oxide particles, in a total amount of from 0.05 to 40 wt%, preferably from 0.1 to 35 wt%, still preferably from 0.5 to 20 wt%, based on the total weight of the composition. If the content is less than 0.05 wt%, the resulting composition is not sufficiently improved in resistance to copper contact, thermal resistance in closed systems, and elimination of mold corrosion, which are the objects of the present invention. On the other hand, if it exceeds 40 wt%, the resulting composition is poor in mechanical properties.

The composition of this invention may contain additives such as antioxidant, thermostabilizer, lubricant, nucleation agent, UV absorber, and coloring agent in ordinary quantities. In addition, the composition of this invention may be blended with other polymers and fillers in an amount that would not hinder the object of the present invention in order to improve the fluidity, crystallinity, thermal resistance, and mechanical properties. If a polymer like polyethylene terephthalate which is poor in thermal resistance is to be incorporated, the quantity should be less than 10% to maintain the thermal resistance of the composition.

It goes without saying that if such additives, polymers, and fillers to be incorporated contain the abovementioned metal components, which are the essential components of the composition of this invention, care must be taken that the total quantity of these components should be in the above-mentioned range (0.05 to 40 wt%).

In the case where the above-mentioned metal components, which are the essential components of the composition of this invention, are dispersed in the composition in the form of fine particles of simple substances, compounds, or mixtures thereof, particles smaller than 1 µm in particle diameter should account for at least 10% by number of the total particles.

The term "sheetlike material" as used in this specification means flat plastic materials thinner than about 5 mm and denotes generally plastic materials called film, sheet, or plate.

The resin composition of this invention exhibits its improved resistance to copper contact and thermal resistance in closed systems when formed into sheetlike materials (particularly biaxially oriented film).

The biaxially oriented film of poly-p-phenylene sulfide of this invention should preferably contain the 10 above-mentioned metal components in an amount of 0.1 to 35 wt%, preferably 0.5 to 20 wt%, based on the weight of the film.

The PPS constituting the biaxially oriented film should preferably have a melt viscosity of 50 to 3,000 Pa.sec when measured at a temperature of 300°C and a shear rate of 200 sec<sup>-1</sup>.

The biaxially oriented film is not specifically limited with respect to the degree of orientation, but the 15 degree of orientation should preferably be such that the tensile strength in the longitudinal and transverse directions is at least 127.4 Mpa. Usually, the tensile strength does not exceed 294 Mpa.

Biaxially oriented films manufactured from a polymer containing a metal oxide are usually poorer in mechanical properties, particularly elongation, than those manufactured from a polymer containing no metal oxides. It is to be noted that such deterioration of mechanical properties does not occur in the biaxially oriented film of poly-p-phenylene sulfide containing zinc oxide and lead oxide according to this invention. This suggests that zinc oxide and lead oxide are particularly effective.

It is not elucidated yet why the PPS resin composition and PPS sheetlike material of this invention containing specific metal oxides are superior in resistance to copper contact, thermal resistance in closed systems, and extending of the life of molds. It is presumed that these specific metal oxides prevent a very small quantity of decomposition products released from PPS resin at high temperatures.

The resin composition of this invention is produced in the following manner.

PPS, which is the base polymer of the composition of this invention, is prepared by reacting an alkali metal sulfide and p-dihalogenobenzene in a polar organic solvent under high temperature and high pressure. Preferably, it is prepared by reacting sodium sulfide and p-dichlorobenzene in N-methyl-pyrrolidone or other high-boiling point amide polar solvents. This process is well known, and the above process is not limitative.

If films, fibers, and high impact molded materials are to be made from PPS, the reaction at 230 to 280°C in the presence of polymerization modifiers such as caustic alkali and alkali metal carboxylates is preferable.

The polymerization pressure and polymerization time should be determined properly according to the degree of polymerization desired and the type and quantity of modifiers used.

The PPS thus prepared is incorporated with at least one component selected from zinc oxide particles and lead oxide particles, which are the essential components of the composition of this invention. They may be incorporated in any manner and, typically in the following two manners. First, fine powder composed of the above-mentioned compounds, or mixture thereof is added to the polymerized PPS resin before or after it is washed with water to remove solvent, residual salts, and polymerization modifiers, or after drying that follows such washing. Secondly, the washing is performed with water containing the ions of the above-mentioned elements. The latter is applicable if the compounds containing the essential elements are soluble in water.

The preferable composition of this invention that contains the essential elements in the form of oxides may be prepared by adding the oxides of desired elements in the form of fine powder to PPS before or after it is washed with water or after drying that follows such washing. It is preferable from the standpoint of workability to mix using a mixer such fine powder with PPS which has been dried.

The powdery composition thus prepared is melted and extruded into gut using an extruder and the gut is cooled and cut into pellets. (This step is called "pelletization" hereinafter.) Prior to pelletization, the composition may be mixed with fillers, additives, or polymers according to the intended object of use.

The composition in the form of powder or pellets thus prepared should contain the essential components in an amount of from 0.05 to 40 wt%. If the content is less than 0.05 wt%, the composition is not sufficiently improved in "resistance to copper contact" and "thermal resistance in closed systems" which are the objects of this invention. If the content exceeds 40 wt%, the resulting composition is poor in mechanical strength.

The PPS sheetlike material of this invention is usually prepared by supplying the resin composition prepared as above to an extruder or other apparatus for melt extrusion, melting it at 290 to 360°C, extruding the melt from slit die, and quenching the extrudate by, for example, casting it around a rotating drum. The PPS sheetlike material thus prepared may further undergo heat treatment for crystallization so that the thermal resistance and mechanical properties are improved as required.

The biaxially oriented film is prepared by stretching the substantially amorphous unstretched sheet 2.0 to 4.5 times in the longitudinal and transverse directions at 85 to 105°C. Stretching may be accomplished by successive biaxial stretching by rolls and a tenter, simultaneous biaxial stretching by a tenter, tube

stretching, and other methods. The successive biaxial stretching by rolls and a tenter is most favorable in the production efficiency and quality.

The resin composition of this invention having the above-mentioned constitution is extremely improved in "resistance to copper contact", "thermal resistance in closed systems", and "corrosion of mold" in injection molding as compared with conventional poly-p-phenylene sulfide resin compositions. The "plastic materials" prepared from the PPS resin composition can be used in any application areas because of the extremely high reliability.

It is surprising to note that the resin composition of this invention is much superior in thermal resistance in ordinary open systems to conventional poly-p-phenylene sulfide resin compositions and the "plastic materials" prepared from the resin composition of this invention can be used continuously at higher temperatures than those from conventional PPS resin compositions.

The sheetlike materials of this invention is extremely improved in "resistance to copper contact" and "thermal resistance in closed systems", without degrading the superior characteristic properties of the conventional poly-p-phenylene sulfide sheetlike materials. The PPS sheetlike materials are extremely reliable and can meet any use conditions.

The biaxially oriented films of this invention are particularly superior in these properties and are useful for wire covering and other electrical insulating applications.

In this specification, the property values of the resin compositions and plastic materials prepared therefrom were defined, measured, and evaluated as follows:

(1) Measuring method of quantity of metal components in composition

The essential metal components in the composition were determined by ashing a sample of the composition using a low-temperature plasma asher, dissolving the ash in acids, converting the solutions into aqueous solutions of nitrates, and finally subjecting the aqueous solutions to ICPES (inductively coupled plasma emission spectrometry).

(2) Apparent melt viscosity

The shear stress  $\tau$ , the apparent shear rate  $\dot{\gamma}$ , and the apparent viscosity  $\mu$  are defined as follows:

 $\tau = (RP)/(2L)$ 

 $\dot{y} = (4Q)/\pi R^2$ 

 $\mu = \tau/\gamma$ 

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when the molten polymer is extruded using an extrusion plastometer having a capillary tube, L in length and R in radius, at a temperature T, under a pressure of P, and with a throughput of Q. The index of the melt viscosity of the polymer is given by the value  $\mu_0$  at  $\dot{\gamma}=200~\text{sec}^{-1}$  on the curve  $\mu=f(\dot{\gamma})$  obtained by plotting  $\mu$  against  $\dot{\gamma}$ . In this invention, measurements were made using a die having L = 10 mm and R = 0.5 mm at T = 300°C.

(3) Glass transition temperature (T<sub>g</sub>) and melting point (T<sub>m</sub>) of polymer

They were measured according to DSC method.  $T_m$  was defined by the peak temperature of the melting curve.

(4) Tensile strength and ultimate elongation

They were measured according to JIS Z-1702 using an "Instron" type tensile tester.

(5) Thermal resistance and resistance to copper contact

The resin composition is formed into a film, about 50 µm in thickness, and the film is heat-treated at 230°C for 1 min. This test piece is measured for the initial values of tensile strength and ultimate elongation according to the above-mentioned methods.

A. Thermal resistance in open systems

The test specimen is placed in a hot-air oven at 220°C for 96 h or 240 h, and the aged test specimen is measured for tensile strength and ultimate elongation. The thermal resistance in open systems is indicated by the percentage of the measured value to the initial value.

B. Thermal resistance in closed systems

The test specimen (about 100 cm²) is placed in a glass test tube, about 50 cm³ in volume, and the opening of the test tube is melted and closed after reducing pressure to 48 kPa. (360 mm Hg). The test tube containing the test specimen is placed in a hot-air oven at 220°C for 96 h or 240 h, and the aged test specimen is measured for tensile strength and ultimate elongation. The thermal resistance in closed systems is indicated by the percentage or the measured value to the initial value.

C. Resistance to copper contact

The test specimen held between two copper plates is placed in a hot-air oven at 220°C for 96 h or 240 h.

The aged specimen is measured for tensile strength and ultimate elongation. The resistance to copper contact is indicated by the percentage of the measured value to the initial value.

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The present invention is described in detail with reference to the following example.

## Example 1

### (1) Polymerization of PPS

Into an autoclave were changed 32.6 kg of sodium sulfide (250 mol, containing 40 wt% of water of crystallization), 100 g of sodium hydroxide, 18.0 kg of sodium benzoate (125 mol), and 79.2 kg of N-methyl-2-pyrrolidone (abbreviated as NMP hereinafter). The reactants were heated up to 205°C slowly with agitation, and 7.0 liters of distillate containing 6.9 kg of water was removed. To the remaining mixture were added 37.5 kg (255 mol) of 1,4-dichlorobenzene (abbreviated as DCB hereinafter) and 20.0 kg of NMP. The reactants were reacted at 250°C for 3 h. The reaction product was washed 8 times with deionized water prepared by ion-exchange method and then dried at 100°C for 24 h using a vacuum dryer. PPS was obtained in an amount of 23 kg. The PPS was found to have an apparent viscosity of 400 Pa.sec at a shear rate of 200 sec<sup>-1</sup> at 300°C, a T<sub>a</sub> of 90°C, and a T<sub>m</sub> of 280°C.

### (2) Preparation of composition

The PPS powder prepared in the above step (1) was blended, using a supermixer, with fine metal oxide powder as shown in Table 1 in such an amount that the metal component accounts for 1 wt% of the total weight. Thus six kinds of compositions (No. 1 to No. 6, including control) were prepared.

### (3) Preparation of plastic materials

The composition prepared in the above step (2) was pelletized using a twin-screw extruder, each screw being 30 mm in diameter. The resulting pellets were then melted at 310°C by a 30-mm single-screw extruder, the melt was extruded from a 200-mm long linear T-die having a 1-mm gapped die lip, and the extrudate was cooled and solidified by casting by an electro-static pinning method around a metal drum, with the surface kept at 20°C. Thus a sheet, 170 mm in width and 610 µm in thickness, was prepared. This film was biaxially stretched to 3.5 times the original length in both longitudinal and transverse directions simultaneously using a film stretcher (made by T. M. Long Co., Ltd. in U.S.). The stretched film was then heat-treated 230°C for 1 min. at a constant dimension. Thus film samples (Film No. 1 to No. 6), 50 µm in thickness, were prepared.

The film samples were measured for the content of the metal component according to the abovementioned method. It was found that the content in the film corresponded to the quantity added to the composition within the range of measurement error.

### (4) Evaluation

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The results of evaluation of the film samples prepared in the above step (3) are shown in Table 1. It is to be noted from Table 1 that the films prepared from the compositions of this invention containing specific metal elements are improved in "thermal resistance in closed systems" and "resitance to copper contact" as well as "thermal resistance in open systems".

TABLE 1

		Additives						
Fila No.	Compound	Content in number (%) of particles not larger than 1 µm in dia.	Tensile strength (Mpa)	Initial ultimate elongation (%)	Thermal resistance in open systems*	Thermal resistance in closed systems*	Resistance to copper contact*	Remarks
	Zinc oxide	. 41	156.8	46	76	73	71	Example
	Lead oxide	. 28	161.7	46	74	. 82	89	Example
	Titanium dioxide	69	145.04	98	ន	. 52	81	Comparative Example
····	Iron sesqui- oxide	96	146.02	20	20	21	11	Comparative Example
	Antimony sesquioxide	43	143.08	£43	<b>32</b>	. <b>6</b> 2	15	Comparative Example
<b>.</b>	None		151.90	45	2	27	21	Comparative Example

\* Expressed by retention % of elongation after heat treatment at 220°C for 240 h.

## Example 2

(1) Polymerization of PPS

Powdery PPS was prepared in the same manner as in step (1) of Example 1.

5 (2) Preparation of composition

The PPS powder prepared in the above step (1) was blended, using a supermixer, with the zinc oxide used in Example 1 (No. 1 zinc white conforming to JIS prepared by wet process) in varied amounts to prepare seven kinds of compositions containing zinc in different concentrations.

10 (3) Preparation of plastic materials

The composition prepared in the above step (2) was extruded, cast, stretched, and heat-treated as in step (3) of Example 1 to prepare film samples (Film Nos. 7 to 13) having a thickness of 50 µm.

The film samples were measured for the content of zinc according to the above-mentioned method. It was found that the content in the film corresponded to the quantity added to the composition within the range of measurement error.

(4) Evaluation

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The results of evaluation of the film samples are shown in Table 2.

It is to be noted from Table 2 that the object of this invention is achieved only when the content of the 20 metal component is within the specified range.

TABLE 2

Remarks	Comparative Example	Comparative Example	Example	Example	Example	Example	Comparative Example**
Resistance to copper contact*	20	24	62	99	<i>L</i> 9	19	25
Thermal resistance in closed systems*	26	29	99	89	71	. 99	59
Thermal resistance in open systems*	25	99	89	20	72	29	31
Initial ultimate elongation (%)	46	46	47	45	47	42	14
Tensile strength (Mpa)	148.96	151.9	153.86	157.78	154.84	151.9	98.98
Content of zinc (wt%)	0	0.024	0.16	0.80	4.0	32	48
Quantity of zinc oxide added (wt%)	0	0.03	0.2	1.0	5.0	40	09
Film No.	7	<b>ω</b>	တ	10	=	12	13

\* Expressed by retention % of elongation after heat treatment at 220°C for 240 h. \*\*Sample film was stretched 2.5 times (instead of 3.5 times) in both directions to prevent breakage.

### Example 3

(1) Preparation of resin composition

The PPS powder prepared in the above step (1) was incorporated with glass chopped fiber (CS-03-MA411, a product of Asahi Fiberglass Co., Ltd.) in an amount of 40 wt% based on the total weight and fine zinc oxide powder (No. 1 zinc white conforming to JIS prepared by wet process) as used in Example 1 in an amount of 10 wt% based on the total weight. The components were mixed using a supermixer to prepare the composition (No. 14) of this invention.

For comparison, a composition (No. 15) was prepared by incorporating the same PPS powder with the above-mentioned glass chopped fiber in an amount of 40 wt% based on the total weight.

These compounds were pelletized using a twin-screw vented extruder.

(2) Injection molding test

The two kinds of pellets prepared in the above step (1) were formed into 20,000 pieces of 3-mm thick dumbbell specimens using an injection molder, Model N—140 built by The Japan Steel Works, Ltd., under the following conditions. The inside of the mold was inspected for corrosion with naked eyes and using a magnifier.

Molding conditions:

Mold material: SK entirely hardened (HRC 55)

Cylinder temperature: 310°C Injection pressure: 98 MPa Mold temperature: 120°C Molding cycle time: 15 sec

In this test, corrosion was observed in the mold used for the conventional composition (No. 15), whereas no corrosion was observed in the mold used for the composition of this invention (No. 14).

Claims

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- 1. A biaxially oriented poly-p-phenylene sulfide film prepared from a resin composition comprising poly-p-phenylene sulfide as a principal component and 0.05 to 40 wt% based on the total weight of the composition of at least one component selected from fine zinc oxide particles and fine lead oxide particles, wherein particles not larger than one µm in diameter account for at least 10% of the total number of particles.
  - 2. A biaxially oriented poly-p-phenylene sulfide film as claimed in claim 1, wherein the poly-p-phenylene sulfide is composed of at least 95 mol% of recurring units represented by the structural formula

and the apparent melt viscosity of the poly-p-phenylene sulfide is in the range of from 50 to 3,000 Pa.sec when measured at a temperature of 300°C and a shear rate of 200 sec<sup>-1</sup>.

- 3. A biaxially oriented poly-p-phenylene sulfide film as claimed in claim 1, wherein the stretching ratio is 2.0:1 to 4.5:1 in both the longitudinal and the transverse directions.
- 4. A biaxially oriented poly-p-phenylene sulfide film as claimed in claim 1, wherein the tensile strength is not lower than 127.4 Mpa and not higher than 294 Mpa in both the longitudinal and the transverse directions.

Patentansprüche

- 1. Biaxial orientierter Poly-p-phenylensulfidfilm, hergestellt aus einer Harzzusammensetzung die Poly-p-phenylensulfid als Hauptkomponente und 0,05 bis 40 Gew.-%, bezogen auf das Gesamtgewicht der Zusammensetzung, wenigstens einer Komponente aus der Gruppe feiner Zinkoxidteilchen und feiner Bleioxidteilchen enthält, worin die Teilchen im Durchmesser für wenigstens 10% der Gesamtzahl der Teilchen nicht größer als 1 µm sind.
- 2. Blaxial orientierter Poly-p-phenylensulfidfilm nach Anspruch 1, in dem das Polyphenylensulfid aus wenigstens 95 Mol-% sich wiederholender Einheiten der Strukturformel

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aufgebaut ist und die scheinbare Schmelzviskosität des Poly-p-phenylensulfids im Bereich von 50 bis 3000 Pa/sec, gemessen bei einer Temperatur von 300°C und einer Schergeschwindigkeit von 200 sec<sup>-1</sup>, liegt.

- 3. Blaxial orientierter Poly-p-phenylensulfidfilm nach Anspruch 1, worin das Streckverhältnis 2,0:1 bis 4,0:1 sowohl in Längsrichtung als auch in Querrichtung ist.
- 4. Biaxial orientierter Poly-p-phenylensulfidfilm nach Anspruch 1, worin die Zugfestigkeit nicht geringer als 127,4 Mpa und nicht größer als 294 Mpa sowohl in Längsrichtung als auch in Querrichtung ist.

### Revendications

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- 1. Pellicule de sulfure de poly-p-phénylène biaxialement orientée, préparée à partir d'une composition de résine comprenant du sulfure de poly-p-phénylène comme constituant principal et 0,05 à 40% en poids, par rapport au poids total de la composition, d'au moins un constituant choisi parmi des particules d'oxyde de zinc fines et des particules d'oxyde de plomb fines, dans laquelle les particules non supérieures à un micron de diamètre représentent au moins 10% du nombre total de particules.
  2. Pellicule de sulfure de poly-p-phénylène biaxialement orientée selon la revendication 1, dans la composition 1. dans la composition 1. dans la composition 2. Pellicule de sulfure de poly-p-phénylène biaxialement orientée selon la revendication 1. dans la composition 2. Pellicule de sulfure de poly-p-phénylène biaxialement orientée selon la revendication 1. dans la composition 2. Pellicule de sulfure de poly-p-phénylène biaxialement orientée selon la revendication 1. dans la composition 2. Pellicule de sulfure de poly-p-phénylène biaxialement orientée selon la revendication 1. dans la composition 2. Pellicule de sulfure de poly-p-phénylène biaxialement orientée selon la revendication 2.
  - 2. Pellicule de sulfure de poly-p-phénylène biaxialement orientée selon la revendication 1, dans laquelle le sulfure de poly-phénylène se compose d'au moins 95 moles % de motifs répondant à la formule développée

et la viscosité apparente à l'état fondu du sulfure de poly-p-phénylène est dans l'intervalle de 50 à 3 000 Pa.sec lorsqu'elle est mesurée à une température de 300°C et avec une vitesse de cisaillement de 200 sec<sup>-1</sup>.

- 3. Pellicule de sulfure de poly-p-phénylène biaxialement orientée selon la revendication 1, dans laquelle le rapport d'étirage est de 2,0:1 à 4,5:1 tant dans le sens longitudinal que dans le sens transversal.
  - 4. Pellicule de sulfure de poly-p-phénylène biaxialement orientée selon la revendication 1, dans laquelle la résistance à la traction n'est pas inférieure à 127,4 MPa et n'est pas supérieure à 294 MPa tant dans le sens longitudinal que dans le sens transversal.

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